

Order and disorder in the triangular-lattice $t - J - V$ model at 2/3 electron density

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Motivated by the recent discovery of superconductivity in $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$, we use series expansion methods and cluster mean-field theory to study spontaneous charge order, Néel order, ferromagnetic order, dimer order and phase-separation in the triangular-lattice $t - J - V$ model at 2/3 electron density. We find that for $t < 0$, the charge ordered state, with electrons preferentially occupying a honeycomb lattice, is very robust. Quite surprisingly, hopping to the third sublattice can even enhance Néel order. At large negative t and small V , the Nagaoka ferromagnetic state is obtained. For large positive t , charge and Néel order vanish below a critical V , giving rise to an itinerant antiferromagnetically correlated state.

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The recent discovery of superconductivity in the $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ materials has led to considerable theoretical excitement [1–3]. Even though the superconducting transition temperatures in these materials are not as high as in the cuprate based high temperature superconductors, a primary reason for the theoretical excitement is that the underlying lattice-geometry for the spin-1/2 cobalt sites in these materials is triangular. The antiferromagnetic exchange on the triangular-lattice is frustrated and has long been argued to provide the right conditions for exotic spin-physics in terms of resonating valence bonds (RVB) [4,5]. While most recent studies of the triangular-lattice Heisenberg model ground states [6,7] suggest a magnetically ordered phase, RVB physics could still be very important away from half-filling.

The materials $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ have a very complex phase-diagram with doping x and water concentration y that includes superconducting, insulating, charge ordered, and also magnetic and phase separated behavior [8]. Superconductivity is found only in a narrow range of x -values (in the approximate range $1/4 < x < 1/3$) and the transition temperatures (T_c) have dome-like structures with x , with a maximum in the middle and vanishing at the ends [9]. Sodium is primarily believed to be just an electron donor to the cobalt oxide layers, where much of the electronic activity takes place. Thus an interpretation of the T_c variation is that at certain commensurate electron fillings T_c goes to zero and the system becomes a Mott insulator, and superconductivity arises from doping the Mott insulator [1,3].

In this paper, we wish to focus on the origin and nature of insulating behavior at 2/3 electron density, which may be appropriate for Na_xCoO_2 at $x = 1/3$, near the superconducting region. There have been very few numerical studies of $t - J$ or large- U Hubbard models on the triangular-lattice. Quantum Monte Carlo methods are known to suffer from minus sign problems, while the exact diagonalization methods are limited to very small systems. Here we use series expansion methods [10], which are especially appropriate for addressing the Mott insu-

lating behavior. We study the Hamiltonian:

$$H = -t \sum_{\langle ij \rangle} P(c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.})P + J \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4}) + V \sum_{\langle ij \rangle} (1 - n_i)(1 - n_j) \quad (1)$$

The first and second terms are the usual terms for the $t - J$ model, while the last term with coupling V is a nearest-neighbor hole-hole repulsion term. The on-site repulsion is assumed to be infinite and no double occupancy is allowed.

For large V , the electrons will spontaneously charge order, preferentially occupying two of the three sublattices of the triangular lattice. This is equivalent to a fully occupied honeycomb lattice. The honeycomb-lattice Heisenberg model has been studied before [11] and, not surprisingly, the unfrustrated model shows antiferromagnetic Néel order. An interesting question is whether the ability of the electrons to hop to the third sublattice can destabilize the antiferromagnetic order or promote ground states with different symmetries. Our results show that for small V a number of different ground state phases, including charge ordered, Néel ordered, dimerized, ferromagnetic, phase separated and short-range antiferromagnetic phases can arise.

We find that the sign of the hopping matrix element t plays a significant role in the properties of the system. First consider negative t . In this case, charge order is very robust, and for a substantial range of $|t|/J$ values, extends even to $V = 0$. Also increasing $|t|$ causes the magnetization on the honeycomb sites to increase well beyond the value in the pure Heisenberg model. For $V = J = 0$, the Nagaoka ferromagnetic state is the ground state. However, singlet, possibly dimerized phases compete with the Nagaoka state even at very small V and J values. These results support the earlier high temperature expansion results of Koretsune and Ogata [12] who found evidence for substantial low temperature entropy in the $t - J$ models with negative t .

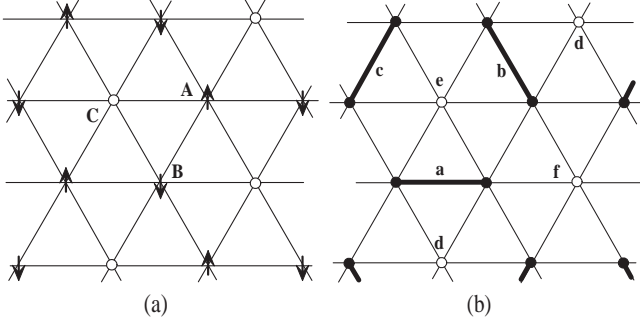


FIG. 1. (a) The Néel ordered state in the Ising limit, (b) the columnar dimer ordered state. The symbols denote the various distinguishable sublattices.

For positive t , hopping reduces Néel and charge order. Below a critical V spontaneous charge and Néel order vanish. From the series expansion one cannot conclusively show that the two vanish simultaneously, but the numerics is consistent with a single transition. At this transition, the symmetry between the three sublattices is restored both with respect to occupancy and hopping. Also, the dimerized and singlet states are never favored for positive t . Thus, for small V and large t , there appears to be a phase transition to an itinerant state with short-range antiferromagnetic correlations.

To perform the Ising expansion, we divided the Hamiltonian into an unperturbed Hamiltonian (H_0) and a perturbation (H_1) as follows,

$$H = H_0 + \lambda H_1 \quad (2)$$

$$H_0 = \sum_{\langle ij \rangle} [J(S_i^z S_j^z - \frac{n_i n_j}{4}) + V(1 - n_i)(1 - n_j)] + U \left[- \sum_{i \in A} S_i^z + \sum_{i \in B} S_i^z + \sum_{i \in C} n_i \right] \quad (3)$$

$$H_1 = \sum_{\langle ij \rangle} [J(S_i^x S_j^x + S_i^y S_j^y) - tP(c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.})P] - U \left[- \sum_{i \in A} S_i^z + \sum_{i \in B} S_i^z + \sum_{i \in C} n_i \right] \quad (4)$$

where λ is an expansion parameter. Note that we have divided the lattice into three sublattices (A , B and C , as shown in Fig. 1(a), The last term in both H_0 and H_1 is a local field term on three sublattices, which can be included to improve convergence. The limits $\lambda = 0$ and $\lambda = 1$ correspond to the Ising model and the original model, respectively. The unperturbed ground state is the usual Néel state (shown in Fig. 1(a)). The Ising series have been calculated for various ground state properties for several ratios of the couplings and (simultaneously) for several values of U to order λ^{11} . The calculation involves a list of 231955 linked clusters (up to 10 sites) for a triangular lattice with 3 sublattices. The series are

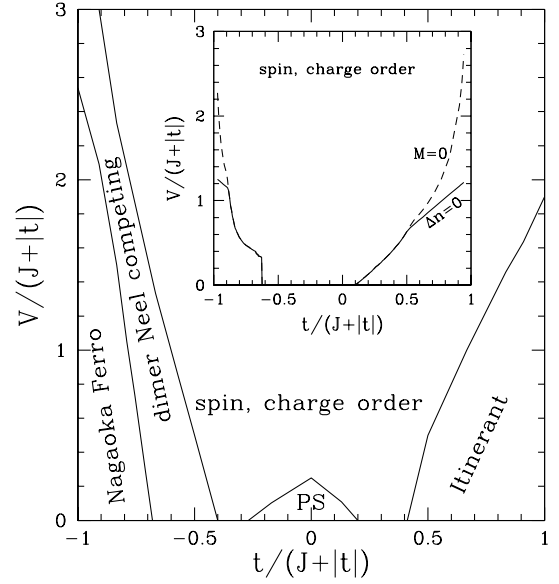


FIG. 2. The phase diagram for the model. PS refers to the fully phase-separated region. Inset shows the phase diagram given by 3-sites cluster mean field calculation.

available on request. Note that $\langle S_i^z \rangle$ on the C sublattice is always zero: this means, in principle, that spin order can occur without charge order.

We also perform a dimer series expansion starting from the columnar dimer pattern as shown in Fig. 1(b), where we take the coupling for bold bonds (the “dimer pairs”) to be J and the coupling for thin bonds (the “free pairs”) to be λJ . The electron hopping amplitude (on all bonds) is also taken to be λt , i.e. we divided the Hamiltonian into an unperturbed Hamiltonian (H_0) and perturbation (H_1) as follows:

$$H = H_0 + \lambda H_1 \quad (5)$$

$$H_0 = J \sum_{\text{dimer pairs}} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4}) + V \sum_{\langle ij \rangle} (1 - n_i)(1 - n_j) + U \sum_{\text{empty sites}} n_i \quad (6)$$

$$H_1 = J \sum_{\text{free pairs}} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{n_i n_j}{4}) - t \sum_{\langle ij \rangle} P(c_{i\sigma}^\dagger c_{j\sigma} + \text{h.c.})P - U \sum_{\text{empty sites}} n_i \quad (7)$$

where again λ is an expansion parameter. The last term in both H_0 and H_1 is a local field term on the empty sites (i.e., the sublattices d , e and f shown in Fig. 1(b)), which can be included to improve convergence. The unperturbed ground state is the product state of dimer singlets. To perform this expansion, we have to divide the lattice into 6 sublattices: 3 for 3 different oriented dimers, and 3 for 3 different empty sites (i.e. the sublattices d , e and f shown in Fig. 1(b)), and this makes it quite diffi-

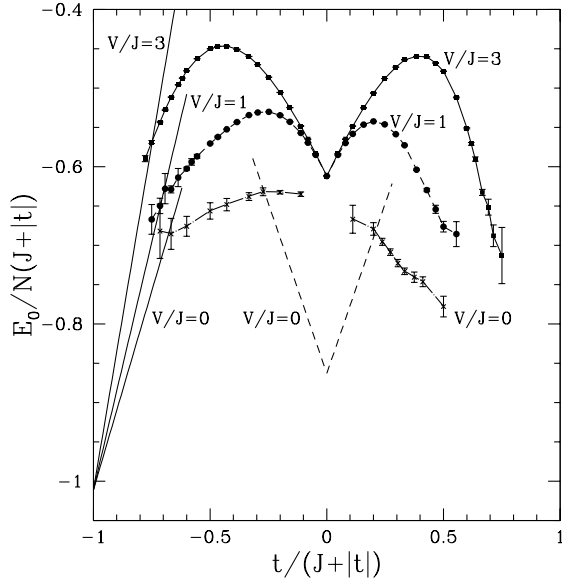


FIG. 3. The ground state energy vs $t/(J + |t|)$ for $V/J = 0, 1, 3$. The points with error bars are the results of the Ising expansion, while the straight lines near $t/(J + |t|) = -1$ are the ground state energies, i.e., $E_0/N = 1.01138t + 0.2481V$, for the ferromagnetic state [15]. The straight lines near $t/(J + |t|) = 0$ are the energies for a fully phase separated state, i.e. $E_0/N = 2Je^{\text{tri}}/3 + V - J/2 = -0.862J + V$, where $e^{\text{tri}} = -0.543$ is the ground state energy per site for the Heisenberg antiferromagnet on the triangular lattice [7].

cult to generate the cluster data for this expansion. The dimer series has been computed to order λ^8 for various ground state properties and for the excitation spectrum, for several ratios of the couplings and (simultaneously) for several values of U [13].

Some of the results are qualitatively confirmed by a cluster mean-field theory. Here a triangle of 3-spins (A , B and C as shown in Fig.1(a)) is considered in the effective field of its surroundings, which favors antiferromagnetic Néel order on two of the sublattices. In other words, we consider the following Hamiltonian

$$H = J(\mathbf{S}_A \cdot \mathbf{S}_B + \mathbf{S}_B \cdot \mathbf{S}_C + \mathbf{S}_C \cdot \mathbf{S}_A) - t(c_{A\sigma}^\dagger c_{B\sigma} + c_{B\sigma}^\dagger c_{C\sigma} + c_{C\sigma}^\dagger c_{A\sigma} + \text{h.c.}) - h_s(S_A^z - S_B^z) + \mu_V \quad (8)$$

where the fields $h_s = 2JM$, $\mu_V = V[\tilde{n}_C(3n_A + 3n_B) + \tilde{n}_A(3n_B + 3n_C) + \tilde{n}_B(3n_A + 3n_C)]$. H needs to be diagonalized with the following self-consistent conditions

$$M = \langle S_A^z \rangle = -\langle S_B^z \rangle, \quad \langle S_C^z \rangle = 0 \quad (9)$$

$$n_A = \langle \tilde{n}_A \rangle = n_B = \langle \tilde{n}_B \rangle, \quad n_C = \langle \tilde{n}_C \rangle \quad (10)$$

The complete phase diagram for the model is shown in Fig. 2. The inset shows the phase diagram obtained by cluster mean-field theory. To obtain the regions of

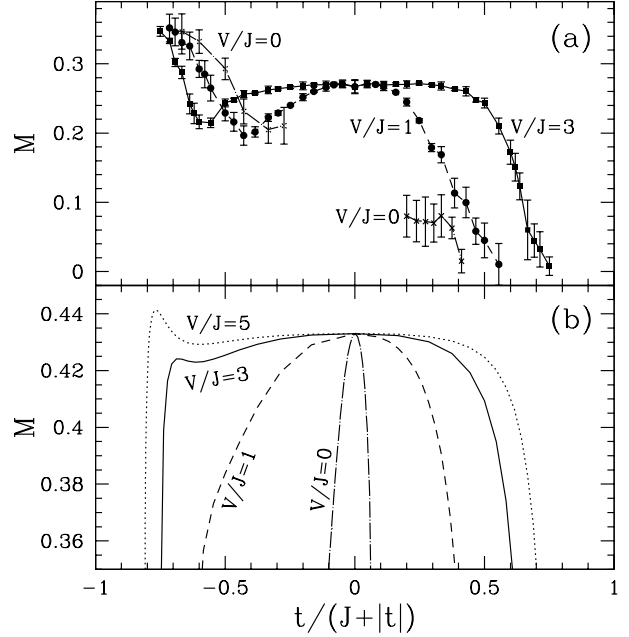


FIG. 4. The magnetization M on sublattice A vs $t/(J + |t|)$ for $V/J = 0, 1, 3$, obtained from Ising expansions (a) and a 3-site cluster mean-field calculation (b).

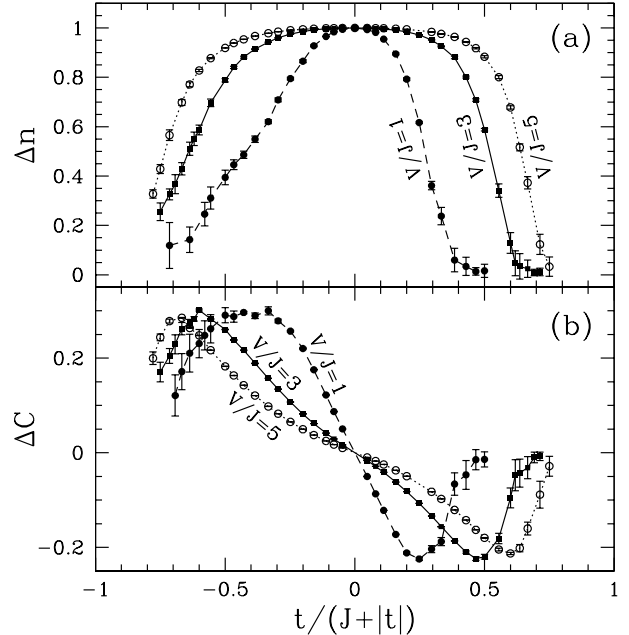


FIG. 5. The difference of electron density on sublattice A (or B) and C $\Delta n = n_A - n_C$ and hopping amplitudes $\Delta C = \langle c_{A\sigma}^\dagger c_{C\sigma} + \text{h.c.} \rangle_0 - \langle c_{A\sigma}^\dagger c_{B\sigma} + \text{h.c.} \rangle_0$ vs $t/(J + |t|)$ for $V/J = 1, 3, 5$, obtained from Ising expansions.

ferromagnetism, we need to compare our calculated energies, with those of the Nagaoka state [14–16]. These are shown in Fig. 3. The phase separated (PS) region is obtained by comparing the calculated energies with those of a phase separated state, with a complete separation of holes and spins. These comparisons are also shown in Fig. 3. Comparison of ground state energies from dimer and Néel expansions leads us to conclude a competing dimer/Néel region adjacent to the Nagaoka phase. This may also reflect the onset of incommensurate spin-correlations, which are not explored here.

The key results of the mean-field theory are that: (i) for negative t , hopping can increase Néel order on the occupied sublattices, and (ii) for positive t and large J charge and spin order vanish simultaneously. For $J = 0$, $t > 0$, this mean-field calculation does not fully capture the Néel order at large V resulting from t/V perturbations. Thus, for small J , charge and spin order vanish at separate critical V 's.

In Fig. 4, we show the variation of the Néel order parameter with hopping. Fig. 4(a) shows the series expansion results and Fig. 4(b) the results of cluster mean-field theory. The qualitative resemblance is clear. For positive t and small V there is a rapid monotonic decrease in Néel order with t . For negative t , increasing $|t|$ leads to a non-monotonic behavior and an increase in the Néel order parameter beyond that of the Heisenberg model. This non-intuitive result is confirmed by the cluster mean-field theory and reflects an interference phenomenon. Fig. 5 shows the restoration of charge order symmetry between the sublattices in the Ising expansions. Fig. 5(a) shows the difference between the site occupations between the initially occupied and unoccupied sublattices. Fig. 5(b) shows the asymmetry in effective nearest-neighbor hopping amplitudes between the different sublattices. The latter shows a non-monotonic behavior with t . However, our results are consistent with the restoration of both types of sublattice symmetry at the same positive t , whereas the symmetry is not restored for negative t . These results suggest that below a critical V and for positive t the system goes into an itinerant antiferromagnetic state with short-range magnetic correlations through a continuous phase transition, whereas for $t < 0$, there is a first order transition to a Nagaoka state.

In conclusion, we have shown in this paper that the triangular-lattice $t - J - V$ model with $2/3$ electron density has a very rich phase diagram that consists of charge ordered, Néel, ferromagnetic, dimerized, phase separated and short-range antiferromagnetically correlated regions. Clearly the full phase-diagram of the triangular-lattice $t - J$ model with doping will be extremely rich, with the possibility of Mott-insulating phases at many commensurate dopings. While it is difficult to directly relate these calculations to the $\text{Na}_x\text{CoO}_2 \cdot y\text{H}_2\text{O}$ materials, our results support the idea that superconductivity in these materials may be viewed as arising from doping a Mott

insulator at $x = 1/3$ provided that $t > 0$ [17,18]. One could further speculate that the water content can change the effective V and bring the system closer to or across the insulating-itinerant phase boundary, thus playing a major role in the onset of superconductivity.

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